Pit Lifetime

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Los Alamos National Laboratory (LANL) and Lawrence Livermore National Laboratory (LLNL) have been tasked by the National Nuclear Security Administration (NNSA) to "provide estimates for predominant pit types" in a Level 1 Milestone Report by September 30, 2006. Results of this assessment by the two nuclear weapons design laboratories could have significant implications for the scope and timing of proposals to restore U.S. capability to manufacture replacement pits. It is therefore important to provide scientifically credible information about pit lifetimes to the decision makers at NNSA. JASON was asked to conduct a comprehensive review of the pit assessment programs of the Laboratories as they approach this Milestone.

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1 EXECUTIVE SUMMARY

JASON reviewed the nearly-completed assessment of primary-stage "pit" lifetimes due to plutonium aging for nuclear weapon systems in the enduring U.S. stockpile. The assessment is being prepared by Los Alamos and Lawrence Livermore National Laboratories in support of NNSA's "Level-1'" milestone to understand possible aging effects in the primary stages of nuclear weapons in the current stockpile and to provide system-specific lifetimes for pits. The joint Laboratory assessment uses the methodology of Quantification of Margins and Uncertainties (QMU) and specifically considers the physical aging effects of plutonium.

We judge that the Los Alamos/Livermore assessment provides a scientifically valid framework for evaluating pit lifetimes. The assessment demonstrates that there is no degradation in performance of primaries of stockpile systems due to plutonium aging that would be cause for near-term concern regarding their safety and reliability. Most primary types have credible minimum lifetimes in excess of 100 years as regards aging of plutonium; those with assessed minimum lifetimes of 100 years or less have clear mitigation paths that are proposed and/or being implemented.

The Laboratories have made significant progress over the past 3-5 years in understanding plutonium aging and pit lifetimes. Their work is based on analyses of archival underground nuclear-explosion testing (UGT) data, laboratory experiments, and computer simulations. As a result of the Los Alamos/Livermore efforts, JASON concludes that there is no evidence from the UGT analyses for plutonium aging mechanisms affecting primary performance on timescales of a century or less in ways that would be detrimental to the enduring stockpile. The detailed experiments and computer simulations performed by the Laboratories to better understand plutonium aging mechanisms and their possible impact on performance of weapons primaries also reduce uncertainties in the expected performance of zero-age pits. The plutonium aging studies are therefore valuable to the overall Stockpile Stewardship program.

JASON identified additional work that should be carried out over the next year or longer to gain a better understanding of relevant plutonium properties and aging phenomena that could affect weapons performance on timescales of a century and beyond.

A more detailed version of this Executive Summary appears in the full (classified) JASON Report.

2 INTRODUCTION

Los Alamos National Laboratory (LANL) and Lawrence Livermore National Laboratory (LLNL) have been tasked by the National Nuclear Security Administration (NNSA) to “provide estimates for predominant pit types” in a Level 1 Milestone Report by September 30, 2006. Results of this assessment by the two nuclear weapons design laboratories could have significant implications for the scope and timing of proposals to restore U.S. capability to manufacture replacement pits. It is therefore important to provide scientifically credible information about pit lifetimes to the decision makers at NNSA. JASON was asked to conduct a comprehensive review of the pit assessment programs of the Laboratories as they approach this Milestone.

Previously, JASON conducted preliminary studies of specific elements of the work of the Laboratories on pit aging. Our studies began with briefings on pit lifetimes presented to JASON by LANL and LLNL in July 2004, briefings in January 2005, a review of the use of underground test (UGT) data in pit lifetime estimates in January 2006, and a followup meeting on the statistical analysis used in April 2006. The findings and recommendations of these earlier phases of the study have been published in classified JASON reports. The final phase of the review was based on briefings that took place in June 2006, two months before the deadline for the Milestone Report. The Laboratory scientists described to JASON their procedures and the majority of their pit lifetime estimates for specific weapons systems.

The purpose of the overall study is to determine whether the research done by the laboratories is adequate to support a reliable pit lifetime assessment for specific systems. Three kinds of research have contributed to the programs of the two Laboratories. The first consists of analysis of results of past underground tests (UGTs) with pits of various ages. Second are studies of the component materials, including experimental and theoretical investigations of the metallurgical properties of Pu containing various combinations of impurities. The experiments involve small-scale (e.g., static compression), medium-scale (e.g., gas-gun dynamic compression), and larger-scale (e.g., hydrotest and sub-critical) experiments. Third are computer simulations of primary performance with model Pu properties varying with age.

JASON was asked by NNSA to consider the following questions:

1. Have the Laboratories identified relevant properties of plutonium, which when varied have significant impact on primary performance? Is this program of research adequate to quantify, bound or, where possible, reduce associated uncertainties? Have appropriate priorities been established?
2. Will the current program of research serve to assess the impact of aging on the properties of plutonium in a reasonably complete and technically sound manner? Will the proposed experiments have the accuracy required to reduce or bound uncertainties? Is the balance amongst activities and program prioritization appropriate?
3. Is the accelerated aging program appropriate and technically sound? Will the planned activities confirm that the accelerated aging samples adequately replicate the properties of naturally aged plutonium and provide a credible extrapolation beyond the age of existing stockpile materials?
4. Are the Laboratories pursuing a program of research for model development and simulation of fundamental plutonium properties and their change with age that will provide useful information in the required time frame?
5. Have the Laboratories provided a scientifically valid and defensible pit lifetime for each of the systems analyzed?
6. Are there areas of uncertainty identified where additional work should be focused?

Questions (1)–(4) were answered in our two previous reports: generally in the affirmative, albeit with a number of recommendations for changing details of the program (to which the Laboratories have been responsive). This report is therefore mainly concerned with questions (5) and (6). Our answers to both of these questions are summarized in the Executive Summary and explained in detail in the body of the report.

3 UNDERGROUND TEST DATA

To adduce evidence for aging, the Laboratories have carried out a detailed examination of the legacy underground test (UGT) data. Though the data are remarkably precise (some critical parameters measured to 1-3%), measurement accuracies were not uniform in time, and accurate errors needed to be established. We conclude that the Laboratories have extracted all possible information regarding pit aging from the UGT data given the uncertainties associated with those data.

4 PLUTONIUM PROPERTIES

Plutonium is a remarkable material. In an electronic sense Pu exists on the knife-edge between localized and delocalized behavior, and these electronic characteristics in part give rise to extensive polymorphism as a function of temperature, pressure, and composition. The δ-phase of Pu stabilized with Ga in the face-centered cubic structure is used in most pits. Pu undergoes radioactive decay and self-irradiation, which causes build-up of Am, U, and Np, and in addition, He bubble formation. These radiation-induced changes lead to complex defects and microstructure. Compounding the problem is the fact that the δ-Pu alloys of interest are unstable under ambient conditions and can partially transform to new phases and phase segregate. Despite these effects there is substantial lattice annealing that counteracts this damage. Indeed, an important finding is that despite the self-irradiation, δ-Pu alloys are remarkably resilient and maintain their integrity (e.g., not undergoing void swelling as discussed below). The question at hand is how changes in physical and chemical properties affect pit performance and on what time scale.

Research on how material properties change with age includes laboratory experiments and computer simulations. Most of the focus has been on Pu and pits. Experiments and calculations on actual and simulated pit materials are combined with experiments on ^{238}Pu-spiked material in accelerated aging experiments. However, the high explosive and other components also need attention. We have reviewed much of the program on pit-material aging in our previous reports, and do not repeat that discussion here. New developments have emerged in the past year, including results published in the open literature.

4.1 Ambient Condition Studies

The best-understood part of Pu aging is the change in its isotopic and elemental composition as unstable isotopes decay. Because half-lives are known very accurately, and relevant cross-sections are generally well known, the contribution of radioactive decay to aging may be calculated with confidence. At early times the dominant contribution is the decay of ^{241}Pu (about 0.5% of pit alloys) with a half-life of 14.4 years to ^{241}Am, which has a lower fission cross-section. At later times, following the depletion of the ^{241}Pu, the rate of decrease resulting from the decay of ^{239}Pu, ^{240}Pu and ^{241}Am is a few times less. If there were no other relevant aging processes these values would themselves imply lifetimes, depending on the margin, of several hundred to over a thousand years.

Surveillance of pits and laboratory experiments on Pu alloys provide direct information on changes in physical and chemical properties with age. Considerable work on density changes in Pu alloys due to aging has been done using volumetric, dilatometric and x-ray diffraction techniques. The results, which were reviewed during the past year, have clarified several inconsistencies. Much of this work involves standard microanalysis, including optical and electron microscopies, and has benefitted from the Enhanced Surveillance and Dynamic Material Properties Campaigns.

The Pu accelerated aging program augments the study of naturally aged Pu. A central question is the extent to which these "artificially" aged samples are representative of "naturally aged" material, given the differences in isotopic composition and heating. A variety of measurements demonstrate qualitative similarities between the two types of material. The samples are held at different ambient temperatures in order to try to match annealing effects. There are also similarities in the density and strength changes. Differences due to the isotopic distribution are well accounted for.

Ga-stabilized δ-Pu is metastable at room temperature. Many of the issues that arise are related to the metastability of the δ-Pu alloy and the nearly 20% volume difference between the δ and α phases. The potential consequences of the thermodynamic metastability for aging of δ-phase alloys have been examined experimentally for both naturally and artificially aged material. Phase decomposition and segregation can occur but the kinetics are slow, with little loss in integrity of the bulk material.

4.2 Equation of State

The equation of state (EOS) is the fundamental thermodynamic relation between the density, pressure, temperature, and composition, and therefore includes the zero-pressure density and compressibility. At least approximately, the measurements between methods and between naturally and accelerated-aged Pu are consistent.

Theoretical calculations are in principle capable of disentangling the separate effects of lattice damage, interstitial and bubble He and chemical impurities and of surveying the entire P–V plane, on and off the Hugoniot. These calculations are generally limited to small simulation cells, while phenomenological calculations are subject to uncertainties in the interatomic potentials. Differential effects of aging may be estimated to useful accuracy even if the absolute accuracy is limited.

There is a need to extend high level computations to the actual performance of aged Pu. LLNL and LANL have both applied large-scale molecular dynamics codes to attempt to simulate the effect of shock compression. This work has been performed on the BlueGene/L supercomputer for various metals. It is important to continue to improve high level calculations on Pu using multiscale modeling approaches, as discussed below.

4.3 Void Swelling

One of the major concerns initially in Pu aging was the possibility of void swelling. Void swelling is a well-known consequence of radiation damage in nuclear reactor material. Because of the potential expansion of material with void swelling, it has been a serious concern. However, there is no empirical evidence for void swelling in aged δ-Pu. This, in itself, is reassuring because in other materials void swelling begins gradually after a finite incubation time, and phenomenological estimates based on these data indicate that any void swelling in δ-Pu will not be significant for several more decades. Even more reassuring is the theoretical expectation that δ-Pu will not undergo void swelling at all. This follows from the fact that the calculated volume increase produced by an interstitial atom in δ-Pu is less (in magnitude) than the calculated volume decrease produced by a vacancy (in materials known to undergo void swelling the inequality is in the opposite direction). This implies that radiation damage will not tend to produce net strain that can be relieved by nucleating a void. Qualitatively, this is expected because δ-Pu has an expanded structure, so that disturbing it will tend to reorganize it in the direction of the denser α phase rather than expanding it. Nucleation of a δ to α transition is prevented by the presence of the stabilizing Ga, which is redistributed by radiation damage so that it is not lost to isolated regions of Pu_3Ga, as would be required for such a phase transformation. In view of the importance of possible void swelling in Pu phases, fundamental studies of the problem should continue, for example using accelerated aged material.

4.4 Strength

Strength is not an equilibrium thermodynamic property and is dependent on many factors. At the outset, it is important to distinguish between different types and measures of strength. These types include compressive yield strength, shear strength, and tensile strength. All are in general strongly dependent on temperature, strain rate, and phase, and can differ for single crystals, polycrystalline aggregates and composites. Thus, the strength of Pu at very high rates of deformation may be different from that observed in static or low strain-rate measurements.

Measurements on Pu at low strain rates show increases in strength with age, either natural or accelerated. This is found both for yield strength (the tensile stress at which irreversible plastic work begins, usually defined at 0.2% strain) and for ultimate tensile strength (the maximum stress achieved before a specimen fails, larger than yield strength because of work hardening). However, these measurements of hardness and strength are either static or quasi-static and performed under ambient conditions, rather than those encountered in the implosion of a pit, and their relevance to nuclear performance is at this time unclear.

We commend the approach taken by the Laboratories for investigating strength in order to obtain a conservative estimate of its effects on lifetimes, but potentially larger effects that might act in the opposite direction have not yet been taken into account. We conclude that the Laboratories have made good progress in identifying possible age-related changes to the dynamic strength of Pu, but there is much work to be done to quantify understanding in the regimes most important for pit performance.

5 LIFETIME METHODOLOGY

5.1 QMU Framework

The laboratories have used the methodology of Quantification of Margins and Uncertainty (QMU) to assess pit lifetimes based on simulations of primary performance. Various metrics for this performance have been established but the key requirement is that the primary must produce sufficient nuclear yield to drive the secondary. It is therefore critical to understand if possible degradation of the pit due to Pu aging will ultimately lead to a failure to ignite the secondary. A large series of UGTs have established that the primary will successfully ignite the secondary provided that the yield is sufficiently large. The basic idea is to compute a ratio of the margin M to the total uncertainty U. The higher this ratio, the higher the level of confidence in the weapon's operation, and, in general, a central goal of Stockpile Stewardship is to continually monitor and assess this ratio and to perform mitigation to increase it should the ratio tend close to 1.

Initial minimum credible lifetime estimates provided by the Laboratories serve to highlight when and where more work is needed for a specific primary system. The non-uniqueness of defining a lifetime for a low margin system is shown by the following. The physics input leads to M and U changing with time as:

$$M(t) = M_0 + St \qquad U(t)^2 = U_0^2 + (\delta S)^2 t^2$$

where we assume that changes are described by a linear slope, S, with an error δS (2σ, to be consistent with U as discussed above), and

$$(\delta S)^2 = \sum_i (\delta S_i)^2$$

Yearly certification demands that $M > U$, so the lifetime T is defined by

$$M(T) = U(T).$$

The determination of lifetime T for then depends on knowing four numbers, M_0, U_0, S, and δS. We have two limiting cases:

1. When the effect of aging is well understood and can be calculated accurately:
$$\delta S \ll S \Rightarrow T \approx \frac{M_o - U_o}{S}$$
2. When the effect of aging has large uncertainty and M_o is not very close to U_o:
$$\delta S \gg S \Rightarrow T \approx \sqrt{\frac{M_o^2 - U_o^2}{(\delta S)^2}}.$$

For systems with low margins, $M_o \approx U_o$ and hence different approaches to error handling will give different answers. These considerations point to the need for continued work on assessment of margins and uncertainties.

6 BEYOND THE LEVEL 1 MILESTONE

The Laboratories have made significant progress toward meeting the Level 1 Milestone, exceeding requirements in some ways, but also identifying work that remains to be done. Although more work is needed, both to provide more complete validation of the lifetime estimates themselves, and to better determine the associated uncertainties and tradeoffs (e.g., mitigation strategies), it is likely that the overall level of effort required is much less than in the past 3–5 years. Another key reason for further work is to gain experience with Pu that has suffered the equivalent of a century or more of aging (i.e., with accelerated aging), thereby allowing an interpolation rather than an extrapolation in estimating performance changes and degradation due to aging. In particular, one wants to know the modes of failure that will be among the first to appear, because these can inform the stockpile surveillance program in order to make it most sensitive to aging-induced degradation.

The following is a listing of recommendations for follow-on studies, with a justification for the need and prioritization (or scheduling) of each recommendation.

1. *Validation through peer review of current estimates of primary-performance lifetimes.* Several systems require more detailed analysis in order to obtain reliable estimates of minimum lifetimes, and their associated uncertainties and tradeoffs. For these systems it is important that each contribution to the lifetime be well understood and validated. In a sense, the issue is not one of accounting for aging but of managing the margins and uncertainties that are already present at zero age, and this is best done by understanding the tradeoffs involved and the consequent mitigation strategies that can be applied. It is our highest-priority recommendation that this effort be completed within a matter of several weeks in order to ensure that

no problems remain unrecognized with the current level of analysis. (We note that this short term recommendation has largely been completed since the writing of this report.)

2. *Primary performance and material strength.* There must be a more detailed understanding of the different types of dynamic (high strain-rate) strengths involved in the weapons codes, and then a more complete understanding of how these strengths vary with aging through relevant experimental and theoretical work. This is fundamentally difficult because strength is not an equilibrium-thermodynamic property, so is not well defined theoretically nor is it always well-defined experimentally. Moreover, the relevant regimes of high pressures, temperatures and strain rates are difficult to access, and the loading-path history and associated kinetics across the material phase diagram are therefore not well determined. New experiments should be carried out on both naturally and artifically aged Pu.

3. *Extended accelerated aging experiments on plutonium.* These include both ongoing study of the current accelerated-aging Pu samples, which are spiked with the rapidly-decaying ^{238}Pu, as well as production of samples that have been aged by alternative means. In all of these cases, the objective is to get the equivalent of multi-century experience on aging phenomena, associated with decay (e.g., radiation damage) as well as with activated processes such as annealing. The latter requires taking sub-samples of accelerated-aged material through various temperature cycles in order to determine how the activated processes have been affected by radioactive decay. This is longer-term (multi-year) work both because time is required for the samples to reach appropriate (equivalent) ages, and because one is looking at effects not likely to influence stockpile weapons for many decades. Nevertheless, such studies are essential in order to validate current understanding, and ensure that no new phenomena lurk unobserved below the surface of existing results, as well as to provide specific predictions of the failure modes to be expected in the stockpile (which in turn inform the surveillance programs on what to look for).

7 FINDINGS AND RECOMMENDATIONS

Our principal findings and recommendations are summarized as follows.

Findings

1. The nuclear weapons design Laboratories have made significant progress in understanding pit aging through improved knowledge of the underlying science and improved techniques for simulating weapons performance. Through their laboratory studies of the materials, including both naturally and artificially aged Pu, and stockpile surveillance activities, the Laboratories have also made significant progress in prioritizing the unresolved questions regarding the aging of stockpile weapons. The labs have also identified key metrics to assess the effects of aging.

2. There is no evidence for void swelling in naturally aged or artificially aged δ-Pu samples over the actual and accelerated time scales examined to date, and good reason to believe it will not occur on time scales of interest, if at all.

3. Systems with large margins will remain so for greater than 100 years with respect to Pu aging. Thus, the issue of Pu aging is secondary to the issue of managing margins.

Recommendations

1. The Level 1 Milestone Report should indicate that the primaries of most weapons system types in the stockpile have credible minimum lifetimes in excess of 100 years and that the intrinsic lifetime of Pu in the pits is greater than a century. Each physical effect on the lifetime of selected systems should be calculated and explicitly reported. The report should emphasize the need to manage margins.

2. Continued work is required beyond the Level 1 Milestone. This includes validating through peer review the current estimates of primary-performance lifetimes for selected primary types, extending accelerated aging experiments on Pu, and determining how aging affects primary performance by way of material strength.

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